

I-1. PROJECT RESEARCHES

Project 8

PR8 Fundamental Research on Decommissioning of Reactor Facility

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OBJECTIVES and RESEARCH SUBJECTS:

The main objective of this research project is to improve the method and the fundamental information on decommissioning of KUR and the Fukushima-Daiichi NPPs. This project consists of seven research subjects from four institutions, as follows.

R4P8-1: Neutron Flux Measurement for Activation Modeling of KUR facility (T. Sano *et al.*, Kindai Univ.)

R4P8-2: Measurement of Activation in the Structure Materials of Reactor for Decommissioning Process (K. Takamiya *et al.*, Kyoto Univ.)

R4P8-3: Fundamental Research about Radioactive Contamination Survey on Nuclear Reactor Facility (S. Fukutani *et al.*, Kyoto Univ.)

R4P8-4: Neutron Capture Cross-Section Measurements with TC-Pn in KUR for Some Nuclides targeted for Decommissioning (S. Nakamura *et al.*, JAEA)

R4P8-5: Identification of Nuclear Material in Spent Fuel (Y. Nauchi *et al.*, Central Research Institute of Electric Power Industry)

R4P8-6: External Neutron Source for Non-Destructive Analysis of Fuel Debris (Y. Nauchi *et al.*, Central Research Institute of Electric Power Industry)

R4P8-7: Study on Non-destructive Analysis Method for Fuel Debris using Neutron Resonance Absorption (J. Hori *et al.*, Kyoto Univ.)

One research project (R4P8-3) could not be reported in this fiscal year since a sampling inspection of the object is under way.

MAIN RESULTS and CONTENTS of this REPORT:

T. Sano *et al.* (R4P8-1) installed an “ERYNGII” neutron detector system around E-4 of the biological shield. The detector system consists of a BF₃ detector, a polyethylene moderator, and a cadmium plate for measuring the epi-thermal neutron flux. The results indicate that the ERYNGII can be used to efficiently observe the epi-thermal neutron flux on the surface of the KUR biological shielding.

S. Fukutani *et al.* (R4P8-3) corrected core samples from the KUR stack made of RC (Reinforced Concrete) and γ -spectrometry by a high-purity Ge detector was per-

formed. As for the inner side sample, no significant artificial nuclide was detected by 200,000 second measurement. In contrast, Cs-137 was detected in the outer side sample with enough long measuring time. The detected Cs-137 was considered to be from fallout of atmospheric nuclear test in 60s~70s.

S. Nakamura *et al.* (R4P8-4) measured the neutron capture cross-sections of ⁴⁵Sc, ⁶³Cu, ⁶⁴Zn, ¹⁰⁹Ag, ¹¹³In and ¹⁸⁶W, which were important reactions from the viewpoint of clearance level in decommissioning. The activation experiments were performed by using the TC-Pn of KUR. For the cross-section of ⁴⁵Sc, it was found that its evaluation is appropriate. For the other nuclides, their results deviate from the guideline, and therefore it follows that their thermal-neutron capture cross-sections must be modified to match the guideline.

Y. Nauchi *et al.* (R4P8-5) measured the gamma-ray spectrum from the short-lived fission products (FPs) by irradiating a uranium and aluminum alloy sample at the LINAC neutron source facility. They focused on the gamma-ray events out of phase (OOP) of the pulsed source by using a TOF method. In the time region from 20 to 50 ms, the gamma-ray peaks due to the short lived FPs such as ⁹⁰Rb, ^{95,97}Y, and ¹³⁶Te were identified in the gamma-ray pulse-height spectrum. In this work, possibility of spectroscopy for OOP events was explored for measurement of FP from ²³⁵U.

Y. Nauchi *et al.* (R4P8-6) obtained the response of the neutron induced gamma ray spectroscopy (NIGS) for an 2.45 MeV neutrons generated by fusion reactions of deuterium in plasma formed in an inertial electrostatic confinement device (IEC source). Gamma rays from the stacks of sheets of uranium and polyethylene were measured with a BGO detector. As the current conclusion, the IEC neutron source is preferable to detect fission prompt gamma rays. If we want to quantify capture gamma-rays from SS contained in waste with nuclear material, gamma ray shielding from the IEC device itself would be required.

J. Hori *et al.* (R4P8-7) performed the neutron resonance densitometry (NRD) imaging test using natural uranium, minor actinides such as ²³⁷Np and ²⁴³Am, Cd as a neutron absorber. A gas electron multiplier (GEM) detector was used for transmission neutron detection. By using TOF information, small amount of ²³⁷Np behind the Cd sheet was able to be identified. It was found that the NRA imaging technique with the GEM detector will be a useful tool for screening the fuel debris.

PR8-1 Neutron Flux Measurement for Activation Modeling of KUR facility

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INTRODUCTION: For the decommissioning of the KUR, it is important to evaluate the amount of radioactivation in the generated waste. In particular, it is important to evaluate the radioactivation in the equipments around the biological shield and the neutron tubes. In general, the evaluation of the activation dose is mainly based on numerical calculations (deterministic or probabilistic models), although some part of the activation dose is measured. In numerical calculations, the neutron spectrum and absolute neutron flux are important input information. In recent years, the performance of computers has improved to the point where it is now possible to calculate neutron flux for the entire facility, including buildings, and to evaluate the injected neutron spectrum into facilities and equipment. However, the absolute values often differ by several 10% or more between the calculated and measured values.

Therefore, the purpose was to compare calculated and measured values in this study. In last year's study, a neutron detector was installed on the surface of the KUR biological shield, and preliminary experiments were conducted to evaluate the measurement time and other factors.

EXPERIMENTS: In this experiment, an EneRgY selective Neutron detector containment device for thermal Group neutron InterruptIon “ERYNGII” neutron detector system [1] was installed to measure the epi-fast neutron flux at the surface around E-4 of the biological shield. The ERYNGII shown in Fig.1 consists of a BF₃ detector, a polyethylene moderator, and a cadmium plate. The ERYNGII cuts off the thermal neutron by cadmium cover and improves the sensitivity to the epi-fast neutron by neutron moderation in the polyethylene. Neutron flux measurements were performed at the KUR thermal power of 1 MW, and the measurement time was 450 sec.

RESULTS: A measured pulsed height spectrum is shown in Fig.2. The wall effect of the counting tube can be observed near 750 ch and 1500 ch, and peaks due to alpha particles and ⁷Li particles can be observed near 2400 ch and 2950 ch, respectively. The total counts over 800 ch, which are the signal due to neutrons, were 60867 counts, and the counting rate was 135.3. These results indicate that the ERYNGII can be used to efficiently observe the epi-fast neutron flux on the surface of the KUR biological shielding.

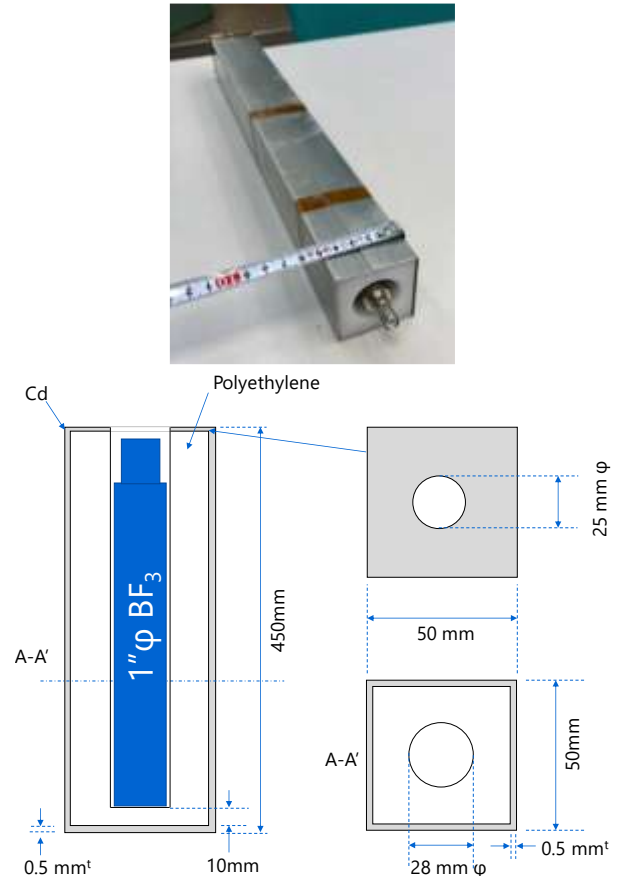


Fig. 1. Photograph and schematic diagram of neutron detector installation to the ERYNGII.

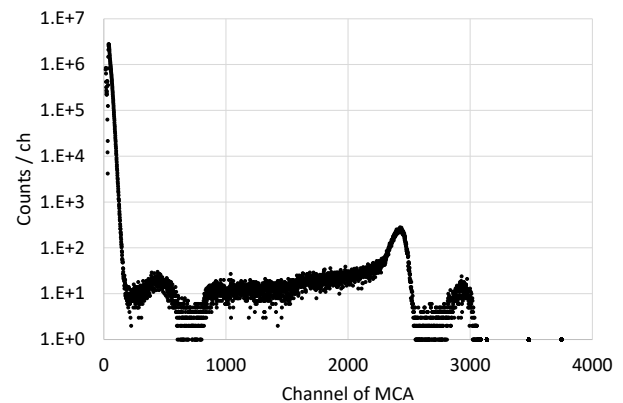


Fig. 2. Measured pulsed height spectrum.

REFERENCES:

- [1] R. Kimura *et al.*, Proc. PHYSOR2022, Pittsburgh, USA, **2824** (2022).

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INTRODUCTION: It is important to survey radioactive contamination for decommission of nuclear reactor facilities. The KUR stack made of RC (Reinforced Concrete) was demolished in 2013, and radioactive contamination of the demolition waste of the stack was surveyed. The stack was built in 1963.

EXPERIMENTS: In demolishing, coring the core was conducted to hang and transport the stack. Core samples were corrected at the time, and were used in this study. One of core samples, shown in Fig. 1, length: 16.2cm, weight: 3440g, was chosen, and about 1mm from the surface of the inner and outer side by using a grinder, respectively. The inner side is contacting side with exhaust gas from the reactor room. The cut sample by the grinder is in powder form, and about 10g of each sample, the inner and the outer, was enclosed into U8 type container, and measured γ -ray by HPGe detector (Ortec GEM 30-70).

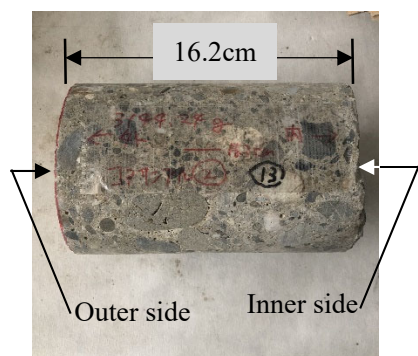


Fig. 1. Core sample from KUR stack.

Inner side is contacting side with exhaust gas, and outer side is faced side with outside air.

RESULTS: γ -spectrometry by HPGe detector was analyzed by the software of Gamma Studio® (SEIKO EG&G). As for the inner side sample, no significant artificial nuclide was detected by 200,000 second measurement. Using the γ -ray peak position of Cs-137 (661.6keV) and that peak area, Activity of Cs-137 was calculated by the Gamma Studio® and standard radiation source manufactured by JRIA (Japan Radioisotope Association). Radio activity and detection limit value of the 661.6 keV peak area for measurement times were shown in Fig. 2. The detection limit gradually decreased in proportion as measurement time increase. However, the radio activity of the 661.6 keV peak area was under the detection limit value at 200,000 second measurement time. In contrast, Cs-137 was detected in the outer side sample. Radio activity and detection limit value of the Cs-137 for measurement times

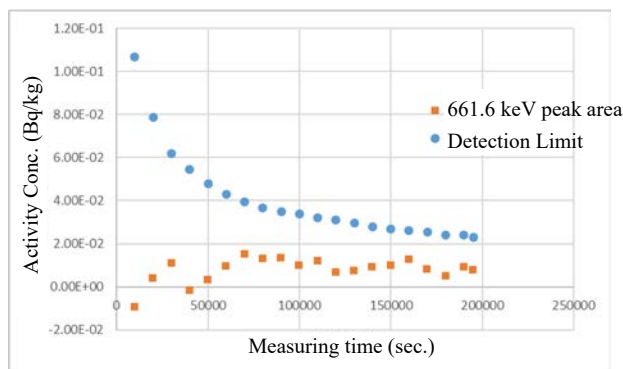


Fig. 2. Changes of activity concentration of 661.6 keV peak area and detection limit with respect to measurement time.

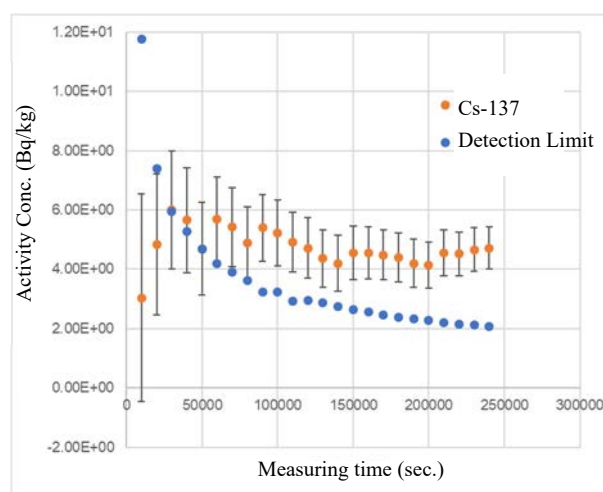


Fig. 3. Changes of activity concentration of Cs-137 and detection limit with respect to measurement time.

were shown in Fig. 3. In short measuring time, 10,000~30,000 second, significant activity of Cs-137 was not detected. But, with enough long measuring time, Cs-137 was detected significantly for detection limit. The detected Cs-137 was considered to be from fallout of atmospheric nuclear test in 60s~70s.

ACKNOWLEDGEMENT: Authors would like to thank members of Technical Staff Office for their contribution to preparation of measuring samples.

PR8-3 Neutron Capture Cross-Section Measurements with TC-Pn in KUR for Some Nuclides targeted for Decommissioning

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INTRODUCTION: Structural materials, piping and equipment of nuclear reactor facilities are generated as radioactive wastes by neutron activation. In the decommissioning of a facility, it becomes necessary to evaluate the amount of radioactivity produced, and this is why accurate neutron capture cross-section data will be required. We have demonstrated that the thermal-neutron capture cross-sections can be accurately derived using the TC-Pn of KUR by measuring ¹⁸¹Ta [1] and ²³⁷Np [2]. It is possible to systematically measure neutron capture cross-sections with use of the TC-Pn irradiation equipment. Consequently, from the viewpoint of clearance level in decommissioning, the present study selected the following nuclides and measured their neutron capture cross-section: ⁴⁵Sc, ⁶³Cu, ⁶⁴Zn, ¹⁰⁹Ag, ¹¹³In and ¹⁸⁶W.

EXPERIMENTS: High-purity metal samples were prepared for neutron irradiation. A gold-aluminum alloy wire, a cobalt foil and a molybdenum foil were used to monitor the neutron flux at an irradiation position. **Figure 1** shows a schematic of irradiation targets. The samples were arranged so as not to interfere with each other as drawn as “Target (a)” in Fig.1. Due to the inner size of the target capsule, the Zn sample was placed at a different position together with a set of neutron monitors as “Target (b)”. To make use of the well-thermalized neutron field, two dummy capsules were sent into the TC-Pn, and followed by the target capsule. The targets were irradiated for 1 hour in 1-MW operation of the KUR.

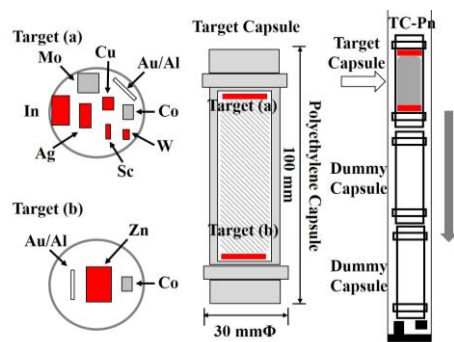


Figure 1. Schematic of irradiation targets.

After irradiation, the target capsule was opened, samples and flux monitors were enclosed in a vinyl bag one by one, and then γ rays emitted from the samples and monitors were measured

with a high-purity Ge detector. The sample was placed at a distance of 100 mm from the front surface of the Ge detector. The γ -ray peak efficiencies of the Ge detector were measured with a ¹⁵²Eu calibration source and a mixed source. In consideration of the radioactivity and half-life of the nuclides, the samples were measured one by one so that sufficient γ -ray peak yields could be obtained.

ANALYSIS and RESULTS: The reaction rates of flux monitors (¹⁹⁷Au, ⁵⁹Co and ⁹⁸Mo) were obtained from their γ -ray yields. Using these reaction rates, the thermal-neutron flux component was derived on the basis of Westcott's convention [3], and found to be $(5.92 \pm 0.10) \times 10^{10}$ n/cm²/sec at the position of Target (a) as shown in **Figure 2**. The thermal flux component is indicated by the guideline in Fig. 2. The obtained reaction rate divided by the evaluated thermal-neutron capture cross-section is also plotted for each nuclide in Fig.2. If the thermal-neutron capture cross-sections are appropriate, the results must fit on the guideline. For the cross-section of ⁴⁵Sc, it was found that its evaluated thermal-neutron capture cross-section is appropriate. So was the cross-section of ⁶⁴Zn of Target (b). For the other nuclides, their results deviate from the guideline, and therefore it follows that their thermal-neutron capture cross-sections must be modified to match the guideline. Considering the uncertainty evaluation, we currently proceed with the analysis toward the derivation of the final results of the thermal-neutron capture cross-sections.

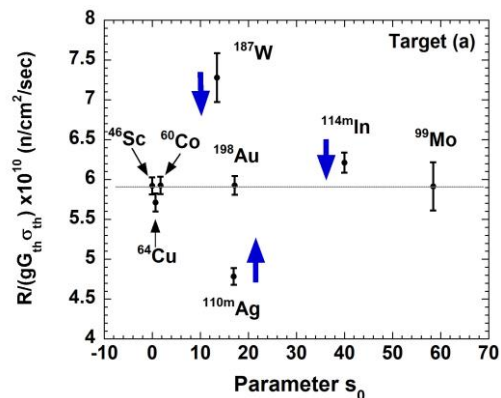


Figure 2. Discrepancies in neutron flux components between the flux monitors and metal samples.

REFERENCES:

- [1] S. Nakamura *et al.*, J. Nucl. Sci. Technol., **58**(10) (2021) 1061.
- [2] S. Nakamura *et al.*, J. Nucl. Sci. Technol., **59**(11) (2022) 1388.
- [3] C.H. Westcott *et al.*, “Proc.2nd Int. Conf. Peaceful Use of Atomic Energy, Geneva”, **16** (1958) 70.

PR8-4 Identification of Nuclear Material in Spent Fuel

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INTRODUCTION: We have conducted γ ray spectroscopy for reactions induced by thermal and resonance energy neutrons at the LINAC neutron source facility in KURNS [1]. In the facility, accelerated pulsed electron is injected onto a tantalum (Ta) target to generate neutrons. The neutrons are moderated in light water surrounding the target. We put a sample at distance 11~12 m from the target and measure the γ ray spectrum from the sample. By measuring time difference between the pulse injection and the γ ray counting, we can determine the energy of neutrons inducing the reactions. The flight time of thermal neutrons (25 meV) from the water to the sample is about 5 ~ 5.5 ms. By setting the pulse frequency 50 Hz, we can obtain the prompt γ ray emission events induced by the thermal and the resonance energy neutrons.

When we measured γ ray spectrum from an Uranium (U) sample of natural ^{235}U enrichment irradiated by neutrons radiated periodically in frequency of 50 Hz, we found γ ray emission in the time region after decay-out of the thermal energy neutron. γ ray spectroscopy for such out of phase (OOP) events might give information of radioactivity of short half-lives induced by neutrons on the phase of the pulsed source. In this work, possibility of spectroscopy for OOP events was explored for measurement of fission products (FP) from ^{235}U .

50Hz MEASUREMENT: An U and aluminum (Al) alloy sample was measured in this work. The ^{235}U enrichment is 97 wt%. The time spectrum of γ ray counting starting from the injection of the pulsed electron is shown in Fig. 1. The spectrum is terminated at the pulse period of 20 ms. The thermal energy peak appears around 3.3 ms. After that, the counting rate decays out and asymptotes to a constant. The spectrum was also measured for a condition where cadmium (Cd) plate is placed upstream of the sample to filtrate out the thermal energy neutrons. By comparison of the count rates with and without the Cd plates in time region from 16 ms to 20 ms, significant count rate of OOP events originated in reaction of ^{235}U induced by the thermal energy neutron are found.

The γ ray pulse height spectra in time regions are shown in Fig. 2. In the thermal energy peaks from 2 ms to 6 ms, count rate is large and continuum components due to ^{235}U fission is significant as well as capture γ rays from ^{27}Al and ^1H . Such prompt components decay with time but $^{27}\text{Al}(n,\gamma)$ and $^1\text{H}(n,\gamma)$ appear until 14 and 16 ms, respectively. Contrarily, 1.778MeV γ ray from decay of ^{28}Al becomes significant as the time increase. ^{28}Al is generated by $^{27}\text{Al}(n,\gamma)$ reaction and its half-life is 2.245 min.

This γ ray is preferable to check stability of pulse height gain.

20Hz MEASUREMENT: To focus on the radioactivity of short-lived FPs free from the prompt γ rays, time region is extended up to 50 ms by changing the pulse frequency to 20 Hz. As shown in Fig. 1, the count rate around the thermal energy peak is reduced to 40 % due to reduction of the beam current compared to the 50 Hz operation. The count rate in the OOP region from 16 to 20ms is also reduced, but the time spectrum becomes stable in time region. The γ ray pulse height spectrum in the time region from 20 to 50ms is shown in Fig. 3. By comparing the measured spectrum to the data in JENDL / FPY & FPD-2011, ^{90}Rb , $^{95,97}\text{Y}$, ^{136}Te , etc. are identified.

REFERENCES:

- [1] Y. Nauchi *et al.*, KURNS pro-gress report 2020 (2021) 93.
- [2] J. Katakura, JAEA-Data/Code 2011-025, 2012.

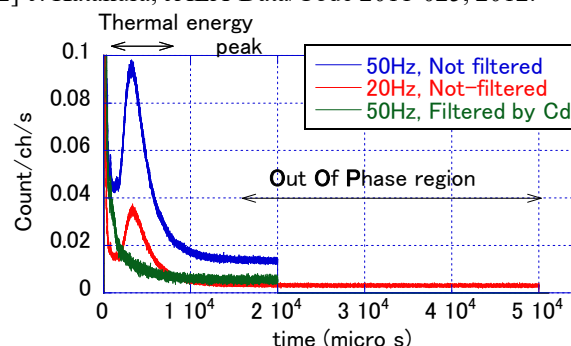


Fig. 1. Time spectrum of γ ray counting from electron pulse injection onto Ta target.

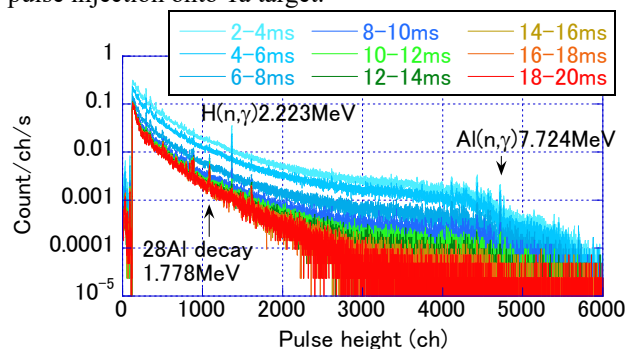


Fig. 2. γ ray pulse height spectrum for time region.

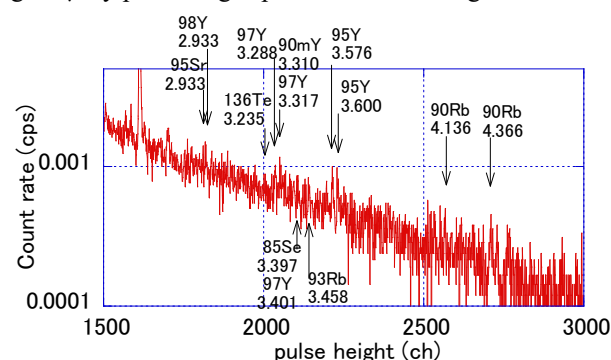


Fig. 3. γ ray pulse spectrum measured for time region from 20 to 50 ms (pulse frequency is 20 Hz).

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INTRODUCTION: Waste retrieved from primary container vessels of unit 1, 2, and 3 of Fukushima dai-ichi nuclear power plant (1F) must be stored safely and efficiently. Unless the amount of nuclear material in the waste is confirmed to be less than a limited value, the waste should be treated caring for criticality safety. Such storage would be a burden for operators. To reduce the burden, we should identify and quantify the nuclear material. For the purpose, the authors have studied the neutron induced gamma ray spectroscopy (NIGS) [1, 2]. By NIGS, the fissile can be identified by measuring fission prompt γ ray. The count rate indicates the products of the amount of nuclear material and the neutron flux in a sub-critical system. For NIGS, we use an external neutron source. One of the candidates for the source is 2.45 MeV neutrons generated by fusion reactions of deuterium induced in plasma formed in an inertial electrostatic confinement device (IEC source). In this study, we obtained response of NIGS for an IEC source in the measurement room of the KUCA facility.

EXPERIMENTS: We used stacks of U sheets of $5.08 \text{ cm} \times 5.08 \text{ cm} \times 0.524 \text{ cm}$ together with polyethylene (PE) sheets of 0.3085 cm in thickness. 20 sets of U and PE sheets were used for a stack and three stacks (U+PE sample) were used for the measurement as shown in Fig. 1. In additional measurement, stacks of U sheets without PE sheets were also used for the sample (U sample). We employed bismuth germinate scintillator (BGO) of 7.62cm in diameter and 7.62cm in thickness. To shield thermal neutron, ^6LiF sheets are placed surrounding the BGO. Polyethylene blocks of 15 cm in thickness was placed between the sample and the BGO to shield the BGO from fast neutrons. The sample was irradiated by neutrons from the IEC source. In the IEC source, deuterium plasma is formed in a cylindrical chamber of radius of 30 cm. The wall of the chamber is made of stainless steel (SS). Lower energy X ray radiated from the plasma is shielded by sheet of lead.

RESULTS: The measured spectra are shown in Fig. 2. The U+PE sample itself radiates considerable amount of γ ray of energy less than 2 MeV, represented by 1.001 MeV γ ray from $^{234\text{m}}\text{Pa}$. The count rate is reduced in energy region greater than 2.615 MeV (green line). Then we focus on the γ ray emission in the energy range greater than 3 MeV in NIGS, as in the case applied for 1F. By the

neutron radiation on PE from the IEC source, $\text{H}(\text{n},\gamma)$ 2.223 MeV γ ray is found (blue, red, black line). Without the samples (blue line) irradiated by neutrons from the IEC source, rather flat spectrum is spread up to 7.7 MeV. That is due to neutron capture reactions in wall of chamber of the IEC source made of SS. That is an intrinsic background when we use the IEC neutron source. The γ ray spectrum from U+PE irradiated by the IEC source (red line) differs from the No Sample case (blue line) in the energy region from 3 to 4.5 MeV. The difference indicates the fission prompt component targeted in this work. This component is also identified when the U sample without PE was used (black dashed line). It is preferable that we can obtain signature of fissile for different neutron moderation condition since the moderation conditions is difficult to be quantified in the waste in 1F. As the current conclusion, the IEC neutron source is preferable to detect fission prompt γ ray since background γ rays in energy range from 3 to 4.5 MeV is rather flat and the count rate is not significant. If we want to quantify capture γ rays from SS contained in waste with nuclear material, γ ray shielding from the IEC device itself would be required.

REFERENCES:

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- [2] T. Shiba *et al.*, Proc. FDR2022, 2022.

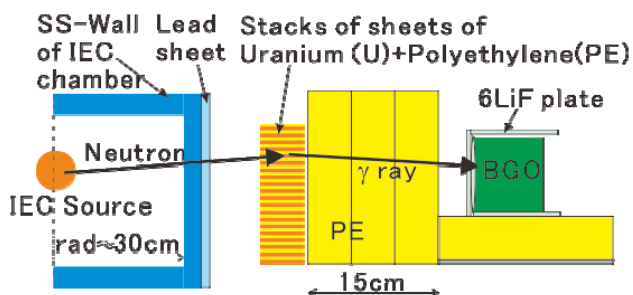


Fig. 1. Schematic view of experimental setup.

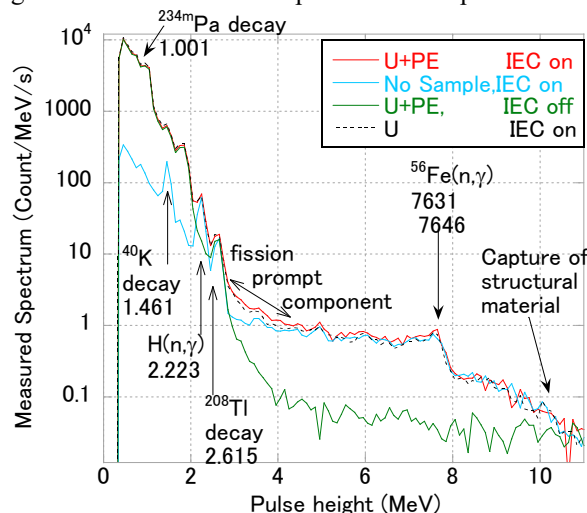


Fig. 2. Pulse height spectrum of γ rays from U+PE or U samples irradiated by neutrons from IEC source.

PR8-6 Study of Non-destructive Analysis Method for Fuel Debris using Neutron Resonance Absorption

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INTRODUCTION: In the units 1 to 3 of Fukushima Daiichi Nuclear Power Plant, the fuel, the metal cladding, and the control rods were melted and large amount were re-solidified in the bottom of the pressure vessel as fuel debris. Characterization of fuel debris is one of the most important issues in the process of the decommissioning. As the fuel debris contains high level radioactive materials such as fissile materials, fission products, and minor actinides, it is difficult to analysis of component directly in the high-dose-rate field. Therefore, the development of non-destructive analysis is necessary for the characterization of fuel debris. We have studied the neutron resonance densitometry (NRD) [1]. In this project research, the NRD technique will be applied to neutron imaging of radioactive material.

EXPERIMENTS: The experiment was performed at the 46-MeV electron linear accelerator in Institute for Integral Radiation and Nuclear Science, Kyoto University. The linac was operated with a repetition rate of 200 Hz, a pulse width of 100 ns, a peak current of about 5 A. The pulsed neutron beam was collimated to 50 mm in diameter. Samples were placed on an aluminum plate of the square of 4 cm. We attached natural uranium foils to the whole plate. Moreover, the sealed minor actinide (MA) samples were placed on the plate. Neptunium-237 and Americium-243 oxide powder were packed in aluminum disk containers, whose inner diameters were 20 mm. The activities of ²³⁷Np and ²⁴³Am were 26 MBq and 950 MBq, respectively. The right half of the plate was covered with a Cd sheet of thickness 0.5 mm. The sample arrangement of the experiment is shown in Fig. 1. The sample plate was placed in the center of beam line at a distance of about 12 m from the neutron source. The beam spot was indicated by a dot line as shown in Fig. 1.

Transmission neutrons were measured by a gas electron multiplier (GEM) detector (THIN-GEM; Bee Beans Technologies Co., Ltd.). We obtained a TOF spectrum for each pixel. The spatial resolution is 0.8 mm and the total area is $10 \times 10 \text{ cm}^2$.

RESULTS: The neutron radiographs obtained by gating on a TOF region are shown in Fig. 2. In the case of thermal neutron region, the part of Cd sheet was only observed and MA samples were not observed. It indicated that small amount of MA cannot be detected with the conventional neutron imaging. On the other hands, the ²³⁷Np sample was identified by gating on the resonance

region of ²³⁷Np. The ²³⁷Np sample behind the Cd sheet was also observed. It is found that the NRA imaging technique has high nuclide identification capacity.

The NRA imaging technique with the GEM detector will be a useful tool for screening the fuel debris.

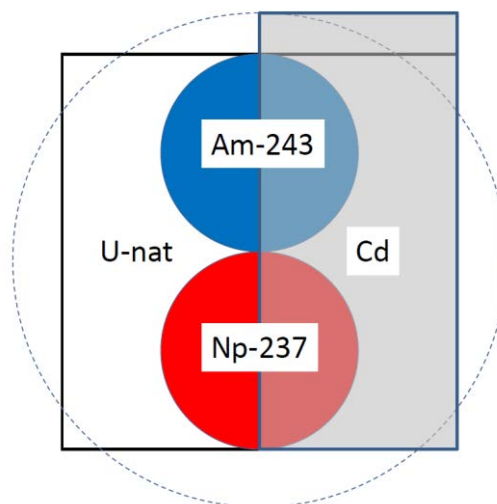


Fig.1 Sample arrangement of U-nat, Am-243, Np-237, Cd for the NRD neutron imaging experiment.

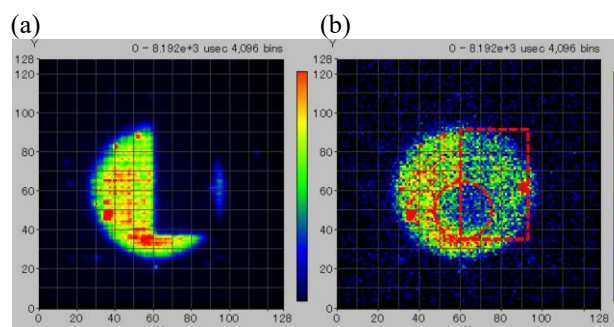


Fig.2 Neutron radiographs obtained by (a) gating on the thermal region and (b) gating on the resonance region of Np-237.

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